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Performance of an Improved Methane-Flow Proportional Counter for TSEE

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An improved methane-flow proportional counter for detecting thermally stimulated exoelectron emission (TSEE), which permits simultaneous measurements of thermoluminescence and TSEE on a single sample, has been developed. The proportional counter is described, and influences on the day-to-day stability and reproducibility of the counter and associated equipment are discussed.

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PERFORMANCE OF AN IMPROVED METHANE-FLOW PROPORTIONAL COUNTER FOR TSEE

INTRODUCTION

The radiation-induced thermoluminescence (TL) of solids has been widely used for personnel dosimetry in nuclear radiation fields. Equipment for reading out the stored thermoluminescent signal has reached quite a high level of sophistication, and several commercial systems are available for rapidly processing TL dosimeter badges [1]. Some researchers have begun recently to focus attention on the phenomenon of exoelectron emission (EE), in which low-energy electrons are emitted from the surface of a radiation-damaged crystal [2]. In particular, thermally stimulated exoelectron emission (TSEE) seems a promising basis for designing new and useful dosimeters for fast neutrons [3].

Detection of low-energy exoelectrons (about 1 eV) requires that the sample be placed inside a windowless gas-flow Geiger or proportional counter, or in vacuum with an electron multiplier. Only one fairly simple counter for the detection of TSEE is commercially available at present (from Atomika, Munich, Germany), and information about other counters used by various workers is fragmentary. In a previous paper [4] the design and operational parameters were given for a methane-flow proportional counter specifically designed for TSEE. In a continuation of that work, we have constructed an improved version of that counter that can be used to make simultaneous measurements of TL and TSEE from a single sample. Various factors that influence the day-to-day stability and reproducibility of the counter have also been investigated and will be discussed in some detail.

MODIFIED PROPORTIONAL COUNTER

A schematic of the improved version of the TSEE proportional counter is shown in Fig. 1. Much of the design is similar to that of the counter described earlier [4]. The sample is placed in the recess of a graphite cup that rests in a gold-plated copper fixture spun down over the top of an automobile cigarette lighter. This entire assembly is inserted into the bottom of the counter and locked into position with a bayonet fitting. A major modification is made by substituting a commercially available hemispherical top* on the counter in place of the cylindrical one used previously. The loop-to-sample distance was maintained at 18 mm, and the position of the loop relative to the chamber top was the same as in the cylindrical chamber. Under these conditions the gas gain characteristics were the same in the two counters. Adding a fused silica window (1 cm in diameter and 1 mm thick) through which to observe any thermoluminescence of the samples also was found to have no influence on the counting characteristics of the chamber.

Note: Manuscript submitted August 1, 1974.

*From an Alpha-Beta-Gamma proportional counter, Model PC-3C, part number NMC-00141, manufactured by Nuclear Measurements Corporation, Indianapolis, Indiana.

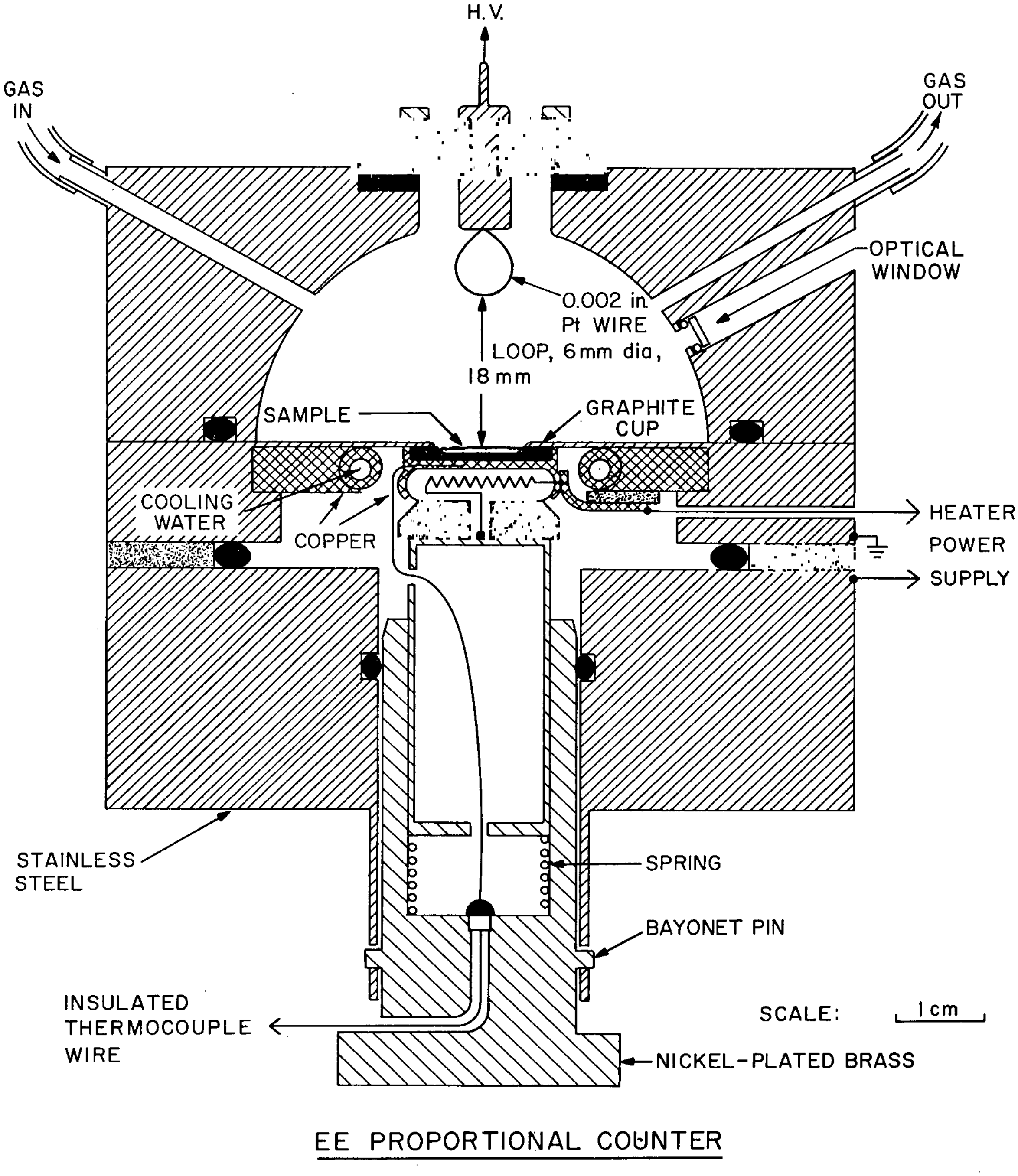

Fig. 1 — Modified methane-flow proportional counter for TSEE

Electrical contact for the heater current is provided by a gold-plated leaf spring pressed against the side of the copper cover on the cigarette lighter. Previously, the leaf spring was fastened directly to the underside of the chamber floor [4]. In the modified design, it is electrically isolated as shown in Fig. 1 so that the current cannot take a parallel path through the lip of the graphite sample dish, leading to possible counting noise and variability in heating rate. Electrical power to the leaf spring is brought in by an insulated wire that passes through a hole in the side of the counter. This hole is sealed with a small amount of putty, not shown in Fig. 1, to prevent the escape of the methane counting gas.

During readout of the samples, a convection column of hot methane gas rises from the sample to the collecting loop. The gas gain in the methane has been found to be temperature dependent, increasing at higher temperatures. While the water-cooled copper coil shown in Fig. 1 prevents the walls of the chamber from heating up, it has little influence on the hot gases rising directly from the sample. The heating of the methane gas during sample readout to 400°C causes great distortion in the pulse-height distribution and a shift in the count-rate plateau [5]. These spurious effects may be diminished by increasing the gas flow through the counter to break up the column of hot gas rising from the sample to the collecting loop. The hemispherical chamber has an advantage over the cylindrical one in this respect because the gas enters and leaves through two vents angled directly at the convection column. Thus the counting gas flows across the convection column and disrupts it more easily than in the cylindrical chamber, where it enters near the floor of the chamber and exits at the top. A gas flow rate of about 100 cm^3/min has been found to eliminate the spurious heating effects in the new hemispherical chamber [5].

A schematic of the EE counting apparatus is shown in Fig. 2. Potentials up to 5000 V can be applied to the collecting loop by a Hamner Model N-4050 power supply. The output from an Ortec Model 109 PC preamplifier (X1 gain setting) was passed through a Model 410 linear amplifier (X5 input attenuation, X9 coarse gain, X2.25 fine gain, 1-μs differentiation and integration times) to a Model 406A single channel analyzer (0.08-V lower discrimination level; 10.0-V upper level). The output was fed simultaneously to a Model 441 linear ratemeter and a Model 430 scaler with a Model 703 overflow register. The output of the ratemeter provided the exoelectron glow curve of the sample being tested, and the scaler showed the total number of counts under the exoelectron glow curve. The counting cycle was started and stopped by a thermocouple amplifier and trip unit consisting of a Brown Electronik recorder whose pen had been replaced with a cam that tripped microswitches mounted above the pen transport carriage. The counting cycle was usually started at 50°C and stopped at 400° or 500°C.

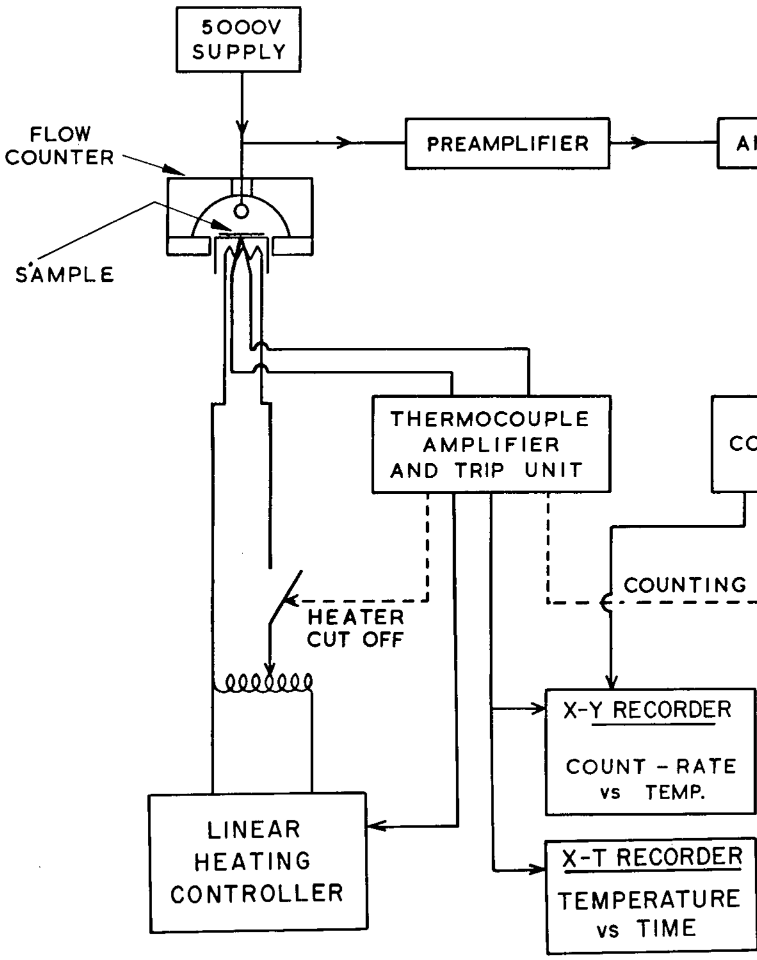

Fig. 2 — Schematic of EE counting apparatus

Typical curves of count rate vs collecting loop voltage for the modified proportional counter are shown in Fig. 3 for beta rays from a ^{14}C source and exoelectrons from LiF (TLD-100). The curve for ^{14}C is the same at 10 and 100 cm^3/min methane flow rates, since emission of the beta rays does not require heating the sample and the attendant convection column of hot gas. The LiF (TLD-100) sample requires heat to read out the exoelectrons and thus is susceptible to distortion of the count-rate plateau because of the increase in gas gain with temperature. The exoelectron plateau begins at lower voltages with the 10 cm^3/min gas flow rate because of the additional gain from the hot column of gas. At 100 cm^3/min the plateau begins at higher voltages, as illustrated in Fig. 3. Because of these effects, an accurate gas flowmeter (e.g., Matheson Flowmeter Model 622PB1 with No. 610 metering tube) is useful for standardizing the gas flow rate through the counter from day to day.

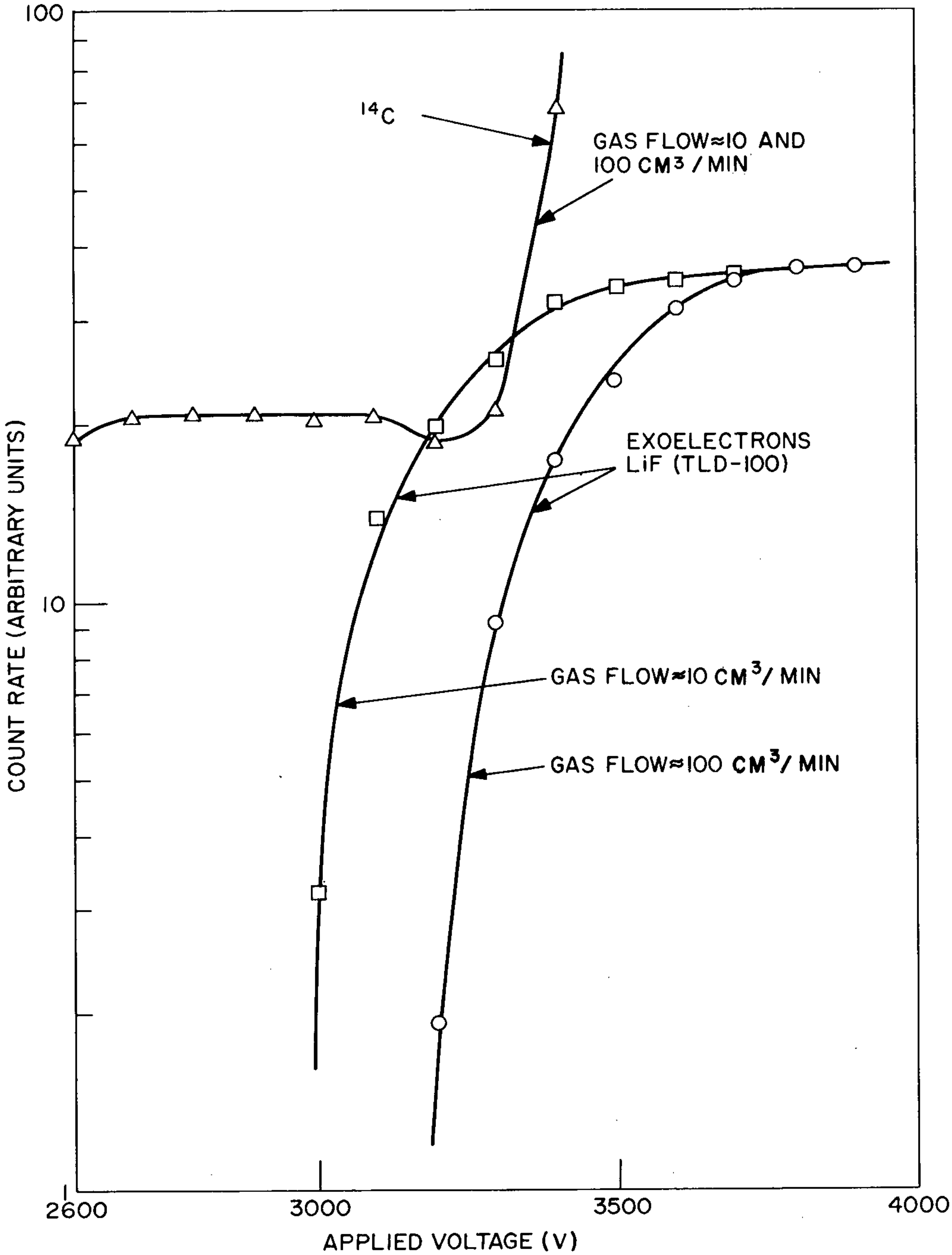

Fig. 3 — Count rate as a function of voltage at different gas flow rates

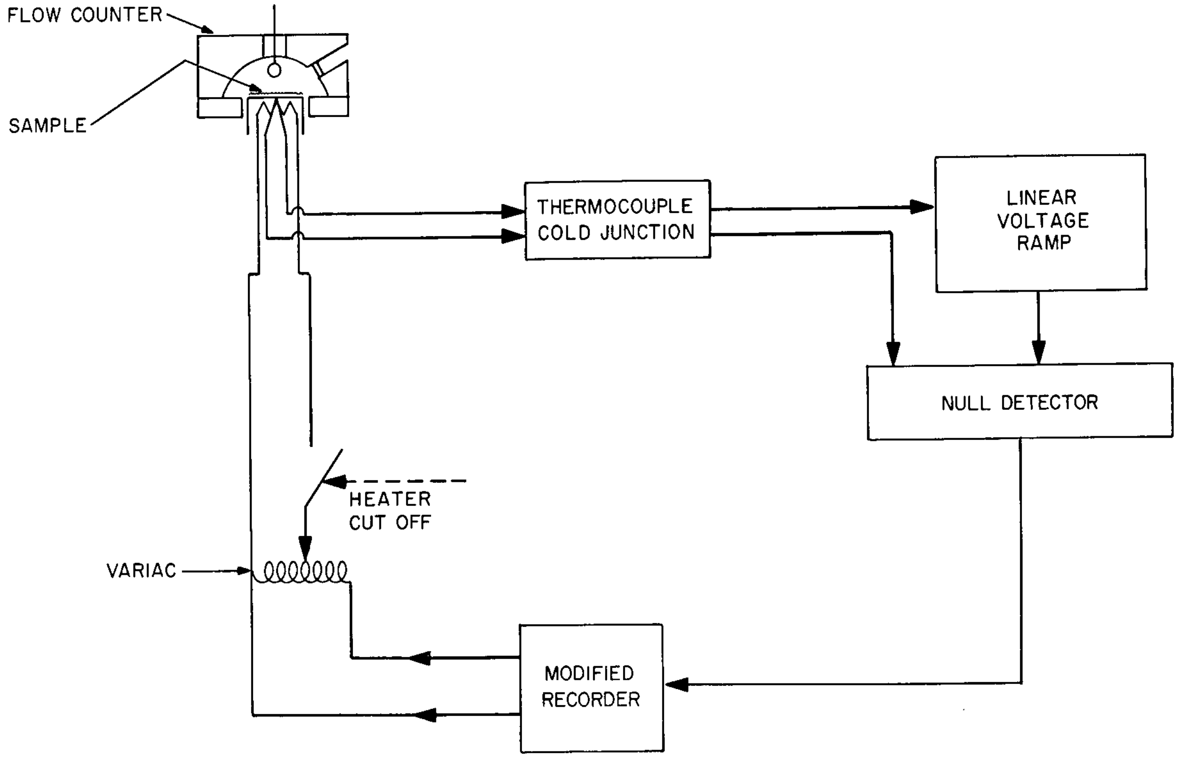

Fig. 4 — Schematic of linear heating control

LINEAR HEATING CONTROL AND TEMPERATURE CALIBRATION

Heating the sample at a linear rate is as desirable in TSEE as it is in TL to avoid distortion of the height and shape of the glow peaks, which would result from nonlinear heating. The linear heating controller indicated in Fig. 2 is shown in more detail in Fig. 4. The sample holder temperature is sensed by a chromel-alumel thermocouple in a 0.5-mm-diameter stainless steel sheath (Omega Engineering No. SCASS-020U-6) used with an automatic cold-junction compensator (Omega Model CJ-K). A Keithley null-detector microvoltmeter (Model 155) senses the imbalance between the control thermocouple and a linear voltage ramp generated by a Hewlett-Packard time base (Model 17108A). The linear ramp operates in three modes: Sweep, Hold, and Reset. The output of the null detector is sensed in turn by another Brown Electronik recorder that has a Variac adjustable autotransformer (110 V, 5 A) mechanically coupled to its pen drive. In this way the autotransformer adjusts the power to the sample heater to maintain a linear heating rate. A stepdown transformer rated at 20 V and 12 A output is placed between the autotransformer and heating element in the proportional counter. Typically, the modified automobile cigarette lighter [6] draws about 8 to 10 A at 10 to 15 V. The thermocouple amplifier and trip unit shown in Fig. 2 cuts off power to the heater when the upper temperature setting is reached, to prevent damage to the heater. In general, the heating control of Fig. 4 maintains the surface temperature of the graphite cup to within ±5°C of the linearly programmed temperature as a function of time during a typical heating cycle from 50° to 400°C.

The 60-cycle pickup noise from the autotransformer was easily filtered out by the Ortec counting system to provide a low count-rate background. This was not the case in early experiments, in which silicon-control rectifiers pulsed by a magnetic amplifier were used. The voltage spikes occurring at the firing points of the silicon control rectifiers were similar in shape to the proportional counter pulses and passed through the amplifier to produce a persistent 60-cycle background that could not be filtered out. A similar problem was encountered by Niewiadomski when a heater supply containing thyristors was used [7].

The surface temperature of the graphite sample cup shown in Fig. 1 was measured in an auxiliary experiment in which the loop assembly was removed and a 0.008-cm-diameter chromel-alumel thermocouple was placed in contact with the center of the cup. Various high-temperature thermocouple cements were investigated to provide good thermal contact between the thermocouple and the graphite cup, but in general they did not provide the good thermal conductivity required for quick response to rapidly changing temperatures during a typical heating cycle. Much better results were obtained by using a small piece of Indalloy solder No. 8 (The Indium Corporation of America, Utica, New York), which melted at 38°C to provide thermal contact between the thermocouple and graphite cup.

The temperature of the upper surface of the graphite was found to be less than that of the control thermocouple embedded in the copper block surrounding the heating element in a steady-state measurement. For a steady-state measurement the voltage ramp was stopped at a particular point for a few minutes, until the temperature difference between the control thermocouple and surface of the graphite reached equilibrium. As shown in Fig. 5, the surface temperature was about 20% lower than the control temperature in the steady state. This is not surprising, since the control thermocouple is between the heater and the surface of the graphite cup. In addition to this temperature difference, the upper surface of the graphite was found to lag behind the control temperature in "dynamic" measurements made during linear heating cycles at 2.5°/s and 5°C/s. Figure 5 indicates that this "dynamic" lag amounts to about 10°C and is more or less constant. The nonlinear heating cycle shown in Fig. 5 was similar to the one without feedback used previously by Attix [4]. In the latter case the temperature lag was most severe early in the cycle and diminished at the end of the cycle, where the heating rate had slowed enough to permit the temperature to approach its steady-state calibration value. A comparison of the TL peak positions measured for various dosimetry phosphors in the apparatus described here [8] with the thermal-quenching work of Gorbics et al. [9] indicates that the measured temperatures are in excellent agreement in the 50°-to-200°C range and within 20°C in the 200°-to-400°C range.

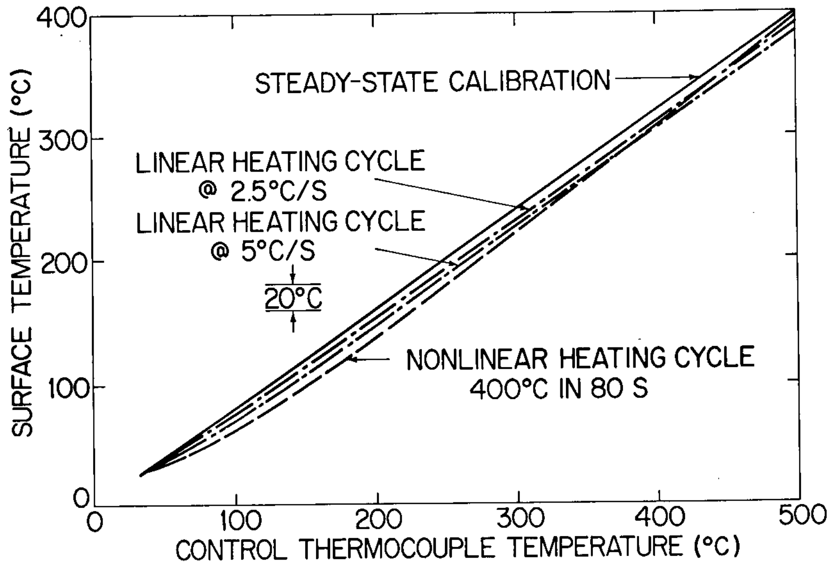

Fig. 5 — Surface temperature of graphite sample cup as a function of control thermocouple temperature for various heating cycles

TECHNIQUES FOR TL AND OPTICALLY STIMULATED EE MEASUREMENTS

A schematic of the thermoluminescence measuring apparatus is shown in Fig. 6. An EMI Model 6094S photomultiplier with a 1-cm-diameter cathode views the sample through the fused silica window. The amount of light exiting through the window is about one-third of that seen if the loop assembly is removed and the photomultiplier is placed over the hole in the top of the chamber. The photomultiplier tube is run at about 950 V (Fluke Model 404M power supply) and is operated in a Jarrell-Ash Model 83-055 PM cooled housing that lowers the dark current to a totally negligible level. A Corning 4303 blue-green filter attenuates the heat signal relative to that of the thermoluminescence light. The photomultiplier output current is amplified with a Keithley Model 410 micro-microammeter, and the total charge is measured with an Elcor Model A308C current integrator. Overall constancy checks of the TL measuring system are made from day to day using a radioluminescent source and the Elcor current integrator. The TSEE count rate and TL current are measured simultaneously during heating of a sample using the apparatus of Figs. 2 and 6 and are recorded on a Honeywell Model 540 two-pen XYY′ recorder. This was found to be especially useful in comparing the TSEE and TL from a given material, since the simultaneous measurement of both outputs during a single heating run eliminates the possibility that sample-to-sample or run-to-run variations in the TL or TSEE output or the heating rate might invalidate the intercomparison [8].

Exoelectron emission may also be stimulated by optical excitation, as has been demonstrated by Petrescu and others [2]. The optical excitation apparatus used with the

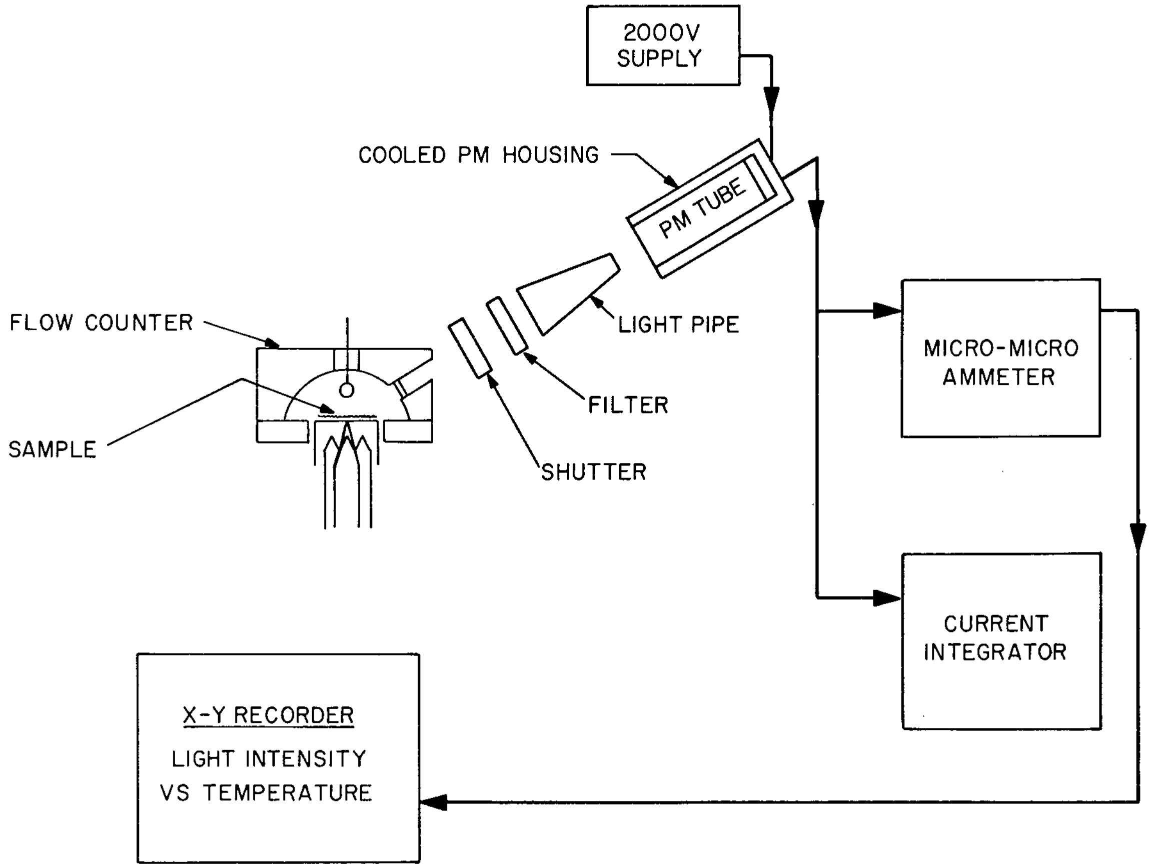

Fig. 6 — Schematic of thermoluminescence measuring apparatus

modified proportional counter is shown in Fig. 7. Light from a xenon arc lamp is passed through a Bausch & Lomb high-intensity monochrometer with a 200-700 nm grating, is collimated, and enters the proportional counter through the fused silica window. The wavelength of the light impinging on the sample may be varied by driving the monochrometer grating with a 1-# to 0.1-rpm reversible variable speed gearmotor (R.M.S. Motor Corporation, catalog no. BG5PlC4BK).

FACTORS AFFECTING DAY-TO-DAY REPRODUCIBILITY

Significant differences have been observed in the intensities and positions of TSEE peaks by various researchers working on the same nominally pure materials. In fact, major changes as a function of time have been observed, apparently caused by changes in surface conditions of the samples as well as in the material itself [8]. The possibility that some of the differences observed were due to the type of counter or counting gas used in the readout was investigated briefly. A comparison of TSEE curves was made between the NRL proportional counter and the Geiger counter [2] used by K. Becker

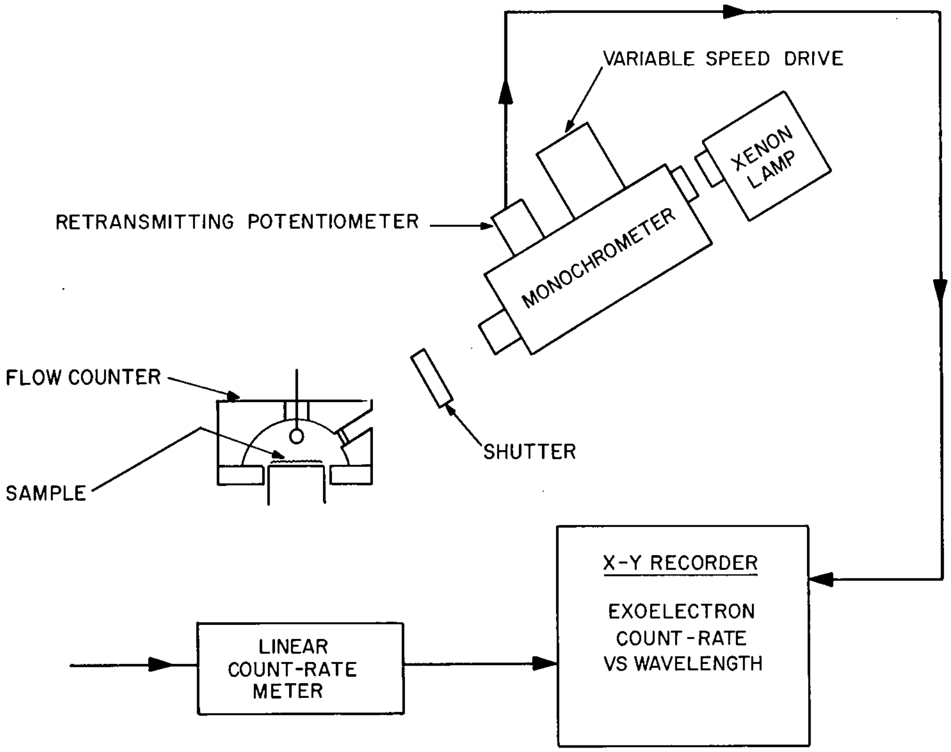

Fig. 7 — Schematic of optical excitation apparatus

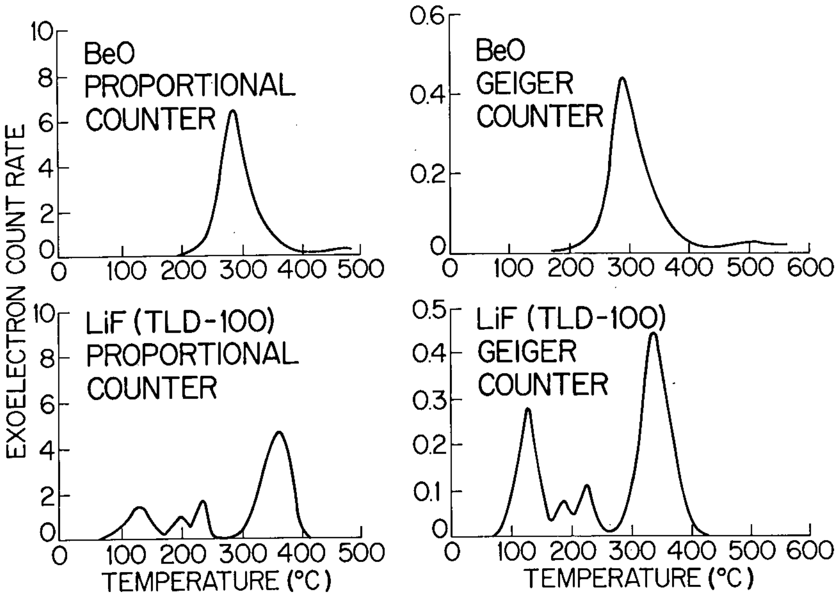

Fig. 8 — TSEE glow curves for BeO and LiF (TLD-100) measured in the NRL proportional counter and the Oak Ridge Geiger counter

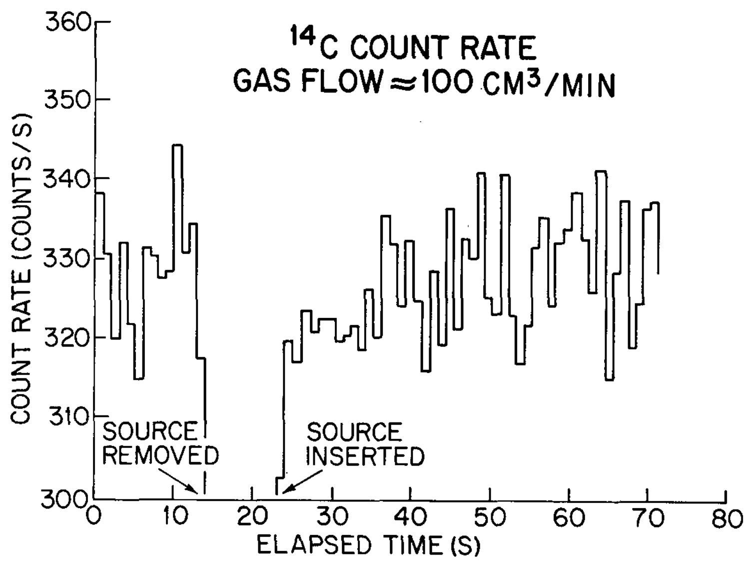

Fig. 9 — Time for proportional counter to reach equilibrium count rate at 100 cm^3/min gas flow rate

at the Oak Ridge Laboratory. Samples of LiF (TLD-100) and BeO, both mixed with graphite, were prepared at Oak Ridge and read out in their Geiger counter, using 99.05% He + 0.95% isobutane as the flow gas. The samples were then hand carried to NRL and measured in the NRL methane-flow proportional counter. The results are shown in Fig. 8. There are no significant differences in glow-curve shape between the Oak Ridge and NRL results. Thus, the type of counter and the counting gas used seem to have little effect on the TSEE curve shape.

It was found, however, that the NRL proportional counter was about 10 times as sensitive as the Oak Ridge Geiger counter. This was probably because the Oak Ridge sample holder was deeply recessed below the floor of the counter. Thus, lines of force from the collecting wire probably terminated on the walls of the recess and did not aid the exoelectrons in their escape from the sample surface. A recent modification of the Oak Ridge counter involving a small positive repelling potential on the sample holder has helped ameliorate this problem, and a higher counter sensitivity to exoelectrons has been achieved [10].

Small amounts of air are inadvertently injected into the NRL proportional counter while changing samples. It was found earlier that this incidental injection of air had little effect on counting efficiency when the counting gas was pure methane, and that the counter quickly came to equilibrium when the sample was inserted [4]. This has been investigated further in the modified proportional counter, using a ^{14}C beta-ray source and an Ortec Model 434 digital ratemeter that integrated the counts over 1-s intervals. Figure 9 shows how long the counter took to come to equilibrium when the methane gas-flow rate was about 100 cm^3/min. At zero elapsed time the equilibrium count rate was being

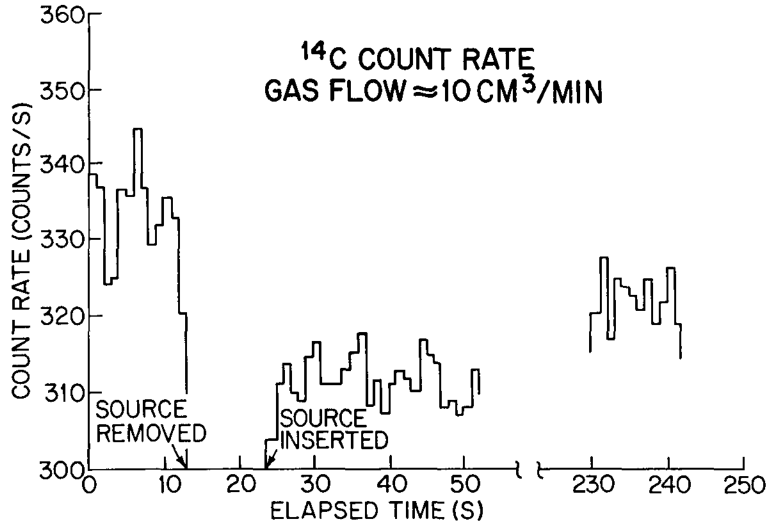

Fig. 10 — Time for proportional counter to reach an equilibrium count rate at 10 cm^3/min gas flow rate

sampled by the ratemeter with the ^{14}C source in the counter. At about 15 s the source was removed and then quickly reinserted. The counter returned to the equilibrium value within about 20 s after reinsertion of the source. The recovery of the counter was much less rapid at lower gas flow rates, as shown in Fig. 10, where a flow rate of about 10 cm^3/min was used. The sequence of events was the same as in Fig. 9, but the counter did not recover to the 330-counts/s (cps) equilibrium value very quickly. The count rate was about 310 cps at 30 s after reinsertion, and an additional 220 s were required before 330 cps was reached. Thus the time to reach full equilibrium can be a problem at low gas flow rates if one tries to make rapid measurements with a precision of better than 10%. Thus it is probably best to operate the counter at a 100-cm^3/min methane flow rate for this reason also. Since the TSEE count-rate plateau is also influenced rather strongly by the flow rate (Fig. 3), an accurate gas-flow meter should be used.

A convenient way to monitor the long-term stability of the proportional counter and associated electronics was found to be the use of a BeO ceramic disk* (Thermalox 995) as an exoelectron-emitting standard. The disk is irradiated with fixed doses of ^{90}Sr beta rays and read out several times each day. It is stored in an air-filled desiccator between readings. The response of such a BeO ceramic disk over a period of several months [11] is shown in Fig. 11. The standard deviation of the day-to-day averages is 4.2%, and inspection of Fig. 11 indicates that the overall sensitivity of the system on any particular day falls within about ±5% of the average. Note that this assumes that the number of exoelectrons emitted by the BeO is constant as a function of time. The use of such an exoelectron-emitting standard has been found to be a much more sensitive check of the overall performance than the ^{14}C beta source used previously, since, as shown in Fig. 3, the exoelectron count-rate plateau lies at much higher voltages than the ^{14}C plateau at high gas flow rates.

*Brush Beryllium Company, Ceramics and Non-Metallic Products Dept., Elmore, Ohio.

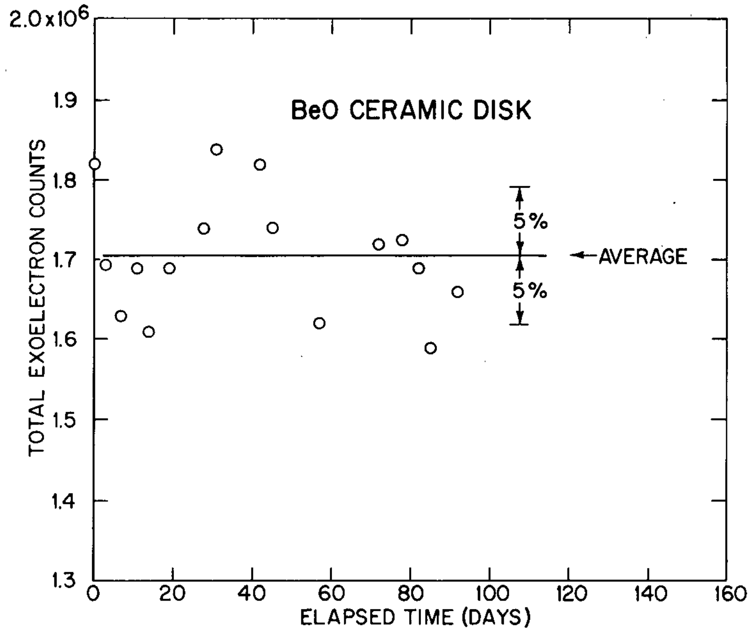

Fig. 11 — Total EE counts from BeO ceramic disk (Thermalox 995) standard as a function of time

While the proportional counter and amplifiers usually maintained the constancy shown in Fig. 11, sudden fluctuations in overall sensitivity have been observed occasionally. A common cause of decreased gain is small specks of dust on the platinum collecting loop in the counter. These are easily removed by dipping the loop assembly in an ultrasonic water bath. A more difficult problem has been fluctuations in the gain of the Ortec 109PC preamplifier after prolonged use. These fluctuations occur in the weeks immediately before failure of the specially selected field-effect transistor in the input stage. Thus, use of a ceramic BeO exoelectron standard is mandatory if precise measurements are to be made on samples over an extended period.

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